The influence of inelastic scattering on EFTEM images—exemplified at 20 kV for graphene and silicon

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1. Introduction

The standard simulation and interpretation of TEM images are based on the elastic scattering theory. This approximation is sufficient for most thin crystalline objects imaged at voltages larger than about 100 kV. In these cases, phase contrast dominates and the contribution of inelastic scattering to the total contrast is negligibly small. However, due to radiation damage caused by atom displacement at higher accelerating voltages > 100 kV, the use of lower voltages (20–80 kV) becomes necessary as realized within the frame of the SALVE (Sub-Angstrom Low-Voltage Electron microscopy) project [1]. As the voltage drops down, the wavelength increases resulting in a decrease of resolution for a fixed usable aperture angle determined by the aberrations of the imaging system. Thanks to the new generation of C$_c$/C$_e$ corrector [2], transmission electron microscopes can now reach sub-Angstrom resolution down to about 60 kV. The novel SALVE corrector provides a usable aperture angle of about 50 mrad, which is supposed to lower the resolution limit to 1.7 Å at 20 kV and to 1.2 Å at 40 kV [3].

When the accelerating voltage is decreased to as low as 20 kV, all objects are strong scatterers [4]. In addition, the elastic model is not sufficient anymore and inelastic scattering must be taken into account especially for low-Z objects. As a result, the interpretation of the images becomes rather involved. In the case of inelastic scattering, the incident electron changes the initial state of the object to any allowed excited state. The initial wave function $\Psi_0$ of the total system is the product of the wave function $\psi_0$ of the incident electron and the wave function $\phi$ of the ground state of the object. After the scattering, the total wave function of the system does not factorize anymore and adopts the entangled form

$$\Psi = \sum_{j=0}^{\infty} \Psi_j |j\rangle. \tag{1}$$

The interaction of the incident electron with the particles of the object results in a transition of the object state from the ground state $|0\rangle$ to an excited state $|j\rangle$ and a change of the wave function of the incident electron from $\psi_0$ to $\psi_j$. For $j=0$, the scattering process is elastic and

$$\Psi_0 = \psi_0 + \psi_e \tag{2}$$

is the sum of the incident wave $\psi_0$ and the elastically scattered wave $\psi_e$. Inelastic scattering is incorporated into the image simulations by means of the Mixed Dynamic Form Factor (MDFF), introduced by Rose [5,6]. The MDFF accounts for the interference of different scattered partial electron waves. The elastically scattered partial waves can interfere with each other, whereas the partial waves of the inelastically scattered electron can only interfere with each other if they are associated with the same excited object state. A detailed discussion is presented in Section 2.1.

We present model-based image simulations for zero-loss and plasmon-loss filtered images at 20 kV for graphene and silicon based on the mutual coherence approach. In addition, a new approximation for the mixed dynamic form factor is introduced. In our calculation multiple elastic scattering and one inelastic scattering are taken into account. The simulation shows that even the intensity of zero-loss filtered image is attenuated by the interference between inelastically scattered waves. Moreover, the intensity of plasmon-loss filtered images cannot be neglected, either.
2$N^2$ times of computational expenditure compared with the 2D case. The image calculation for thick objects usually involves the multislice algorithm, and the computational task required for the propagation of the 4D array through all the slices will be too time-consuming.

In order to tackle the 4D problem in a computation-efficient way, different methods for the factorization of MDFF or density matrix have been proposed with different applications [7–17, 19–22]. Within the core-loss range where the scattering is highly localized, Stalknecht and Kohl [8] as well as Navidi-Kasmai and Kohl [9] proposed the calculation of the density matrix elements based on the first-order perturbation theory combined with Bloch-wave function. Schattenschneider employed the dipole approximation [7] and this approximation was applied for the calculation of EFTEM images by Verbeeck et al. [10]. Dwyer et al. [11, 12] calculated the density matrix elements for atomic ionization based on the work of Saldin [13]. Lügg et al. [14] computed the bound-state wave based on a relativistic Hartree–Fock model. Löffler et al. [15] introduced the method of matrix diagonalization. For the low-loss range where the inner shell structure is neglected, Müller et al. [16, 17] utilized Bessel functions for the factorization of the MDFF obtained by employing the Raman–Compton approximation [18]. In [19–22] the MDFF was calculated by using precise wave functions. The result applies for all energy-losses, but at the sacrifice of efficiency.

This paper concentrates on the image simulation for the low-loss range and our simulation is based on the multislice mutual coherence method outlined in [16, 17]. In addition, we introduce a new approximation for the MDFF. This approximation keeps the maximum similarity with the MDFF function, obtained by utilizing the Raman–Compton model [18] and the Wentzel model [22] for the atom potential. Our approximation can be applied to different imaging conditions, without loss of computational efficiency.

### 2. Theory

#### 2.1. Coherence and incoherence

Coherence and incoherence is the fact that there is a fixed phase relation between the waves (wavepackets) represented by $\psi_j$, so that the waves (wavepackets) can interfere with each other. Incoherence indicates that there is no fixed phase relation between the waves (wavepackets), and the observable intensity is a summation of the intensities of the waves (wavepackets).

The eigenstates of the object are mutually orthogonal, which leads to

$$\langle m|j\rangle = \delta_{mj}.$$  

Therefore the observable intensity can be written as

$$\psi_i^*\psi_i = \sum_{m=0}^{\infty} \sum_{j=0}^{\infty} \langle m|j\rangle = \sum_{m=0}^{\infty} |\psi_m|^2.$$

Since the intensity $\psi_i^*\psi_i$ is the superposed intensity contributed by each scattered partial wave corresponding to different object eigenstates, the conclusion is that the scattered waves are incoherent with each other as long as the coupled object states are different. The second term implies that the elastically scattered wave $\psi_0$ can interfere with the non-scattered wave $\psi_0$.

For elastic scattering, the incident wave function $\psi_0$ propagates through the object. However, a pure wave function cannot be applied for the description of imaging process involving inelastic waves because inelastic waves are always coupled with the corresponding object states. Rose was inspired by the concept of mutual coherence function applied in optics, and extended its usage to the handling of the wave propagation in the electron microscope [5]. Mutual coherence function accounts for the spatial and temporal interference between the waves which correspond to the same object state. Unlike the pure wave function, the propagation of the mutual coherence function $\Gamma_0$ preserves the object information as well as the amplitude and phase resulting from wave interference.

$$\Gamma_0 = \langle \psi^*(\vec{r}, t) | \psi(\vec{r}, t–\tau) \rangle_f.$$  

Here $\tau$ is the temporal difference between the two waves, and $\vec{r}$ and $\vec{r}$ indicate that the two waves originate from different sources. $T$ represents the time average.

#### 2.2. The concepts applied for the image calculation involving both elastic and inelastic scattering

In order to clarify the concepts utilized for the image calculation involving both elastic and inelastic scattering, the counterparts used for pure elastic scattering are listed in Table 1, exemplified by a thin sample.

**Table 1**

<table>
<thead>
<tr>
<th>Elastic/Inelastic</th>
<th>Pure elastic</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Incoming</strong></td>
<td></td>
</tr>
<tr>
<td>$\Gamma_0 = \langle \psi^*(\vec{r}, t)</td>
<td>\psi(\vec{r}, t–\tau) \rangle_f$ (MCF)</td>
</tr>
<tr>
<td><strong>Target</strong></td>
<td></td>
</tr>
<tr>
<td>$\gamma = \langle \psi^*(\vec{r}, t)</td>
<td>e^{iK \vec{r}} \psi(\vec{r}, t–\tau) \rangle_f$ (MOT)</td>
</tr>
<tr>
<td><strong>Outgoing</strong></td>
<td></td>
</tr>
<tr>
<td>$\Gamma'_0 = \langle \psi(\vec{r}, t)</td>
<td>\psi(\vec{r}) \rangle_f$ (MCF)</td>
</tr>
</tbody>
</table>

The time-averaged phase factor $\langle \gamma \rangle_0$ is proportional to the static projected atomic potential defined as

$$\chi(\vec{r}) = \frac{1}{\hbar v} \sum_{j=1}^{\infty} V_j(\rho, \vec{r}) dz,'$$  

where $\hbar$ is the Planck constant, $v$ is the velocity of the electron and $V_j(\rho, \vec{r})$ is the position-dependent atomic potential of the jth atom.

The Taylor expansion of MOT to the second order of $\chi$ results in Eq. (7), according to [6]

$$\langle \gamma(\vec{r}, \vec{r}', \vec{r}, \tau) \rangle_0 = \exp(m_1(\vec{r}')–m_1(\vec{r})–\frac{1}{2} m_2(\vec{r}') – \frac{1}{2} m_2(\vec{r}) + m_{11}(\vec{r}, \vec{r}', \tau))$$  

where $m_1$, $m_2$, and $m_{11}$ are the linear, quadratic, and cubic coefficients, respectively, of the Taylor expansion of the mutual object transparency function.

The main advantages of using mutual coherence function are that it is based on a general expression of wave propagation and it can be naturally extended to include both elastic and inelastic scattering.

### References

1. Z. Lee et al., The influence of inelastic scattering on EFTEM images—exemplified at 20 kV for graphene and silicon, Ultramicroscopy (2013), http://dx.doi.org/10.1016/j.ultramic.2013.05.020

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Incoming MCF caused by the contribution of the inelastically scattered electrons. Eq. (7) satisfies the optical theorem, indicating that the intensity of the incident MCF is conserved:

\[ I(\rho = \rho^*, \tau = 0) = 1. \] (11)

By assuming an average energy loss \( \Delta E \) and by neglecting the interactions between the atomic electrons, we obtain the expression for the MDFF \( S(K, K', \Delta E) \) of a multi-atom system

\[ S(K, K', \Delta E) = \sum e^{-iK \cdot \rho} \phi(K \cdot \rho) \delta(E - \Delta E) F_i(K - K') \]

\[ - \frac{1}{n!} \left( \sum f_j \phi_j(K \cdot \rho) \right)^2 e^{-iK \cdot \rho} \rho^2 d^2 \rho \]

(12)

which is a sum of the contributions from the single atoms. Here \( \rho_e \) is the positional vector of the \( e \)th atom, \( \phi_j \) is the wave function of the jth electron in the \( e \)th atom at the ground state. The elastic form factor \( F_i(K - K') \) describes the coupling of two waves at the ground state of the \( e \)th atom. The Raman-Compton approximation neglects the exchange and the correlation of the electrons to a certain extent. This approximation has been introduced by Lenz [18] for electron scattering in the case \( K = K' \). For \( K \neq K' \), the Raman-Compton approximation for a single atom with atom number \( Z \), has the form [5]

\[ S_i(K, K', \Delta E) = \left[ F_i(K - K') - \frac{1}{n!} \sum F_i(K - K') F_i(K') \right] \delta(E - \Delta E) \] (13)

and for a multi-atom system

\[ S(K, K', \Delta E) = \sum e^{-iK \cdot \rho} \phi(K \cdot \rho) \delta(E - \Delta E) F_i(K - K') \] (14)

Here the elastic form factor of the \( e \)th atom

\[ F_i(K) = Z \int \phi(K \cdot \rho) e^{-iK \cdot \rho} d^2 \rho \] (15)

is the Fourier transform of the corresponding electron density.

The advantage of Eqs. (13) and (14) is to allow to account for inelastic scattering by the elastic form factor of a single atom. We can replace \( \delta(E - \Delta E) \) in Eq. (14) with the energy distribution \( p(E) \delta(\Delta E) \) obtained by normalizing the EELS spectra: \( \int p(E) \delta(\Delta E) = 1 \). The calculation of \( S(K, K', \Delta E) \) utilizes the EELS spectra of the inelastically scattered electrons. The plasmon contribution is neglected in the single-atom model (Eq. (14)). However, by incorporating the experimental energy loss distribution, we consider the contribution of plasmon excitation with sufficient accuracy, because the delocalization for plasmon energy losses is large compared with the mean distance between the neighbor atoms. Based on Eqs. (9), (10) and (13) one can derive

\[ \mu_2(\rho) = \int_{\text{inel}} \mu_{11}(\rho = \rho^* + \Delta \rho) \, d(\Delta E). \] (16)

The energy distribution function \( p(E) \delta(\Delta E) \) is included in the term \( \mu_{11}(\rho = \rho^* + \Delta \rho) \). The elastic form factor \( F_i(K) \) in Eq. (13) can be obtained by applying Wentzel model [23] which adapts the screened Coulomb potential for a single atom.

\[ F_i(K) = \frac{Z}{1 + K^2 \alpha^2}. \] (17)

where \( \alpha \) is the screening radius with \( \alpha = a_0 \rho_{B}^{-1/3} \) and \( a_0 = 0.0529 \) nm is the Bohr radius. In the case of axial plane-wave illumination, the scattering angle equals the aperture angle \( \theta \). Assuming in addition small energy loss \( \Delta E < E_0 \), we obtain

\[ K^2 \approx k_0^2 (\rho_{B}^2 + \rho^2). \] (18)
respectively. The 4D Fourier transform can thus be turned into the combination of independent 2D Fourier transforms. Correspondingly, according to Eq. (9), \(j_{\gamma j_{11}}\) can also be written as a combination of functions depending on \(\vec{r}^1\) and \(\vec{r}^2\), respectively. In order to factorize \(S(\vec{r}^1, \vec{r}^2; \Delta E)\), we have derived an approximation (Appendix A).

3. Method

In our image simulation, multiple elastic scattering events and one inelastic scattering event are taken into account. The sample is sliced into \(M\) layers, and the probability of inelastic scattering taking place at every layer is the same, therefore the result should be a statistical average. If the inelastic scattering takes place at the \(n\)th layer, then in front of as well as behind the \(n\)th layer the propagation of the mutual coherence function \(\Gamma\) is always assumed to be elastic. The procedure follows the order:

1. Input the initial mutual coherence function: \(\Gamma_0 = \psi_0(\vec{r}^1; \vec{r}^0)\). In the case of coherent axial illumination, \(\psi_0\) is a plane wave.

2. Propagate \(\Gamma_0\) through the first \(n-1\) layers. If inelastic scattering takes place at the first layer, this step can be skipped.

\[\Gamma_{n-1}(\vec{r}^1, \vec{r}^2) = \int \Gamma_{n-2}(\vec{r}^1, \vec{r}^2) \Gamma_{n-1}(\vec{r}^0) \mathcal{P} \, \mathrm{d}\vec{r}^0.\] (22)

Here \(\mathcal{T}\) is the transmission function adopting the form

\[\mathcal{T} = \exp\left( -\frac{\mathbf{k}^2 z}{4 \lambda} \right)\] (23)

and \(\mathcal{P}\) is the Fresnel propagator. The computation of convolution \(\mathcal{P}\) is usually converted into a product of Fourier transforms, when the Fresnel propagator adopts the form:

\[\mathcal{P}(q) = \exp\left( -\frac{q^2 z}{4 \lambda} \right)\] (24)

with \(z\) the slice thickness and \(\lambda\) the wave length of the incident electron wave.

3. Inelastic scattering at the \(n\)th layer. We approximate \(\gamma\) further as shown in Eq. (25). The calculation order in this and the following steps is shown in Fig. 2.

\[\Gamma_n(\vec{r}, \vec{r}', \Delta E) = \Gamma_{n-1}(\vec{r}, \vec{r}') \gamma_n(\vec{r}, \vec{r}', \Delta E)\]

\[\approx \Gamma_{n-1}(\vec{r}, \vec{r}') \gamma_{n-1}(\vec{r}, \vec{r}', \Delta E) \exp\left( -\frac{q^2 z}{4 \lambda} \right)\]

\[\times \left[ b_0 + b_1 \mu_{ij} \right],\] (25)

Here the constant coefficients \(b_0\) and \(b_1\) are determined by the linear fitting of \(e^\mu\) in the vicinity of \(x^\mu\), which satisfies \(e^\mu = b_0 + b_1 \cdot x\). In the \(n\)th layer, one elastic scattering takes place, indicated by the constant \(b_0\) in Eq. (25) and one inelastic scattering takes place, indicated by \(b_1\). The mutual object transparency \(\gamma_n(\vec{r}, \vec{r}', \Delta E)\) is related to \(\gamma_{n-1}(\vec{r}, \vec{r}', \Delta E)\) in Eq. (7) by Fourier transform:

\[\gamma_n(\vec{r}, \vec{r}', \Delta E) = \int \gamma_{n-1}(\vec{r}, \vec{r}', \Delta E) e^{i \Delta E \chi_n} \, \mathrm{d}t.\] (26)

4. Propagate \(\Gamma_n\) elastically through last \(M-n\) layers, similar to Step 2. If inelastic scattering takes place at the last layer, this step can be skipped.

The propagation of the electron through the objective lens

5. The mutual coherence function at the back focal plane is given by

\[J(\vec{q}, \vec{q}', \Delta E) = \mathcal{F}(\Gamma(\vec{r}, \vec{r}', \Delta E)) e^{-i \vec{q} \cdot \vec{r}} e^{-i \vec{q}' \cdot \vec{r}'} \cdot \int \mathcal{P}(q) e^{i \Delta E \chi_n} \, \mathrm{d}t.\] (27)

The diffraction pattern (DP) is

\[DP = \int J(\vec{q}, \vec{q}', \Delta E) \, \mathrm{d}\Delta E.\] (28)

The intensity \(I\) at the image plane is given by

\[I(\vec{q}) = \mathcal{P}(q) \int J(\vec{q}, \vec{q}', \Delta E) W(\Delta E) \, \mathrm{d}\Delta E.\] (29)

Here \(A(q)\) and \(A(q')\) are the aperture functions, \(W(\Delta E)\) is the energy window, \(\chi_e\) is the chromatic aberration.

\[\chi_e(q) = \frac{\pi C_1 q^2 \Delta E}{E_0},\] (30)

Table 2

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Graphene</th>
<th>Si(110)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample thickness (Å)</td>
<td>3.35</td>
<td>167 ± 10</td>
</tr>
<tr>
<td>No. of atomic layers</td>
<td>1</td>
<td>88</td>
</tr>
<tr>
<td>Sampling rate (Å/pixel)</td>
<td>0.01</td>
<td>0.02</td>
</tr>
<tr>
<td>Cc (mm)</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Cu (nm)</td>
<td>-2</td>
<td>0</td>
</tr>
<tr>
<td>Δf (Å)</td>
<td>42</td>
<td>178</td>
</tr>
</tbody>
</table>

Fig. 3. The absorption factor \(\mu_s\) calculated for the monolayer graphene at 20 kV. As can be seen, the factor \(\mu_s\) for graphene at 20 kV is close to a constant. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
where $C_c$ is the chromatic aberration coefficient. The geometrical aberration $x_g$ is given by

$$x_g(q) = \pi (2\Delta f q^2 + \frac{3}{2}C_s q^4),$$

where $\Delta f$ is the defocus and $C_s$ is the 3rd-order spherical aberration coefficient.

The general multislice procedure is summarized in Fig. 1. Case A denotes pure elastic scattering with absorption, corresponding to the constant $b_0$ in Eq. (25). The image calculated for Case A is the zero-loss filtered image. Case B denotes the inelastic scattering event at the $n$th layer, corresponding to the term $b_1/u_{11}$ in Eq. (25). In front of and behind this layer the propagation is always elastic. The image calculated for Case B is the low-loss filtered image in our case.

4. Results

Graphene and Si(110) are chosen to illustrate the influence of inelastic scattering on an one atom thin sample (graphene) and on a classical thin sample (17 nm silicon), respectively. We have simulated EFTEM images for graphene and Si(110) at 20 kV with parallel illumination. The corresponding aberration parameters providing the maximum atom contrast are determined by the procedure described in [4]. In both cases, the chromatic aberration coefficient $C_c$ is set to zero. The energy distribution $p(\Delta E)$ (Eq. (13)) is obtained from experimental EELS spectra for graphene and Si(110), respectively. The parameters for the calculations are listed in Table 2. The sample thickness of Si(110) is determined by the experimental EELS spectra with log-ratio method [24] to $16.7 \pm 1$ nm, for graphene the thickness of one single layer is $1.23$. The images calculated for graphene and Si(110) are shown in Fig. 4. The normalized zero-loss peak; (b) the zero-loss filtered image without absorption; (c) the zero-loss peak extracted from the normalized EELS spectra; (d) the zero-loss filtered image with absorption; (e) The plasmon peak; (f) the plasmon-loss filtered image; (g) the line profiles marked in the image (b), (d) and (f). The y-axis shows the intensity and the x-axis shows distance in pixels. Sample thickness: 3.35 Å. Aberration parameters: $C_c=0$, $C_s=\pm 2 \mu m$ and $\Delta f = 42$ Å. Scale bar: 2 Å. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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0.335 nm. For both samples the thickness is sufficiently small that multiple inelastic scattering can be neglected.

The usable aperture provided by the latest $C_i/C_c$—corrector, which has been realized on the SALVE II microscope is 50 mrad.

With the new corrector the point resolution at 20 kV can reach as high as 1.7 Å. For Si(110), extra calculations were made for an usable aperture as large as 86 mrad, which is necessary for resolving the dumbbell structure in Si(110) requiring a point resolution around 1 Å.

Experimental energy-loss spectra of both Si(110) and freestanding single-layer graphene were recorded using the SALVE I prototype microscope, which is a Zeiss-Libra-based transmission electron microscope optimized and operated at 20 kV. The microscope is equipped with a monochromator and an in-column Omega-type energy-filter giving an energy resolution of 0.16 eV for graphene and 0.23 eV for Si, respectively [3]. Energy-loss spectra were recorded in image-mode with a collection semi-angle of 7.7 mrad for Si(110) and 10 mrad for graphene determined by the objective aperture. The entrance aperture of the energy filter limited the illuminated sample area to less than 200 nm in diameter.

4.1. Graphene

The factor $\mu_2$ was calculated based on Eqs. (9) and (16), with the MDFF $S_{\langle K, K' \rangle}$ derived with the help of the Wentzel model (Eq. (21)). The part of EELS spectra utilized for the calculation of $\mu_2$ is shown in Fig. 4e).

The influence of the absorption factor $\mu_2$ can be seen from Eq. (25)—the intensity of the zero-loss filtered image is attenuated by the factor of $e^{-\mu_2}$. As one can see from Fig. 3, the absorption factor $\mu_2$ for graphene at 20 kV is close to a constant (check the colorbar values). Therefore we have $e^{-\mu_2} = e^{-0.298} = 0.74$, based on which one can predict that the zero-loss filtered image intensity is decreased to 74% compared with the pure elastic calculation.

Another reason for the different intensity of the zero-loss filtered images calculated based on the method considering inelastic scattering and calculated based on pure elastic scattering is the factor $b_0$ in Eq. (25). In the case of graphene at 20 kV, the coefficients $b_0$ and $b_1$ are fitted based on the value of $(\mu_2)$ which lies within the range [0,0.5]. The linear fitting of $\exp(x)$ within the range results in $b_0=0.97$ and $b_1=1.29$, respectively; while for pure elastic scattering we have always $b_0 = 1$, $b_1 = 0$ and $\mu_2 = 0$.

For graphene at 20 kV, the factor $b_0$ decreases the image intensity about 3%. According to the integration of the EELS spectra in Fig. 4c and (e), only 0.65% of the electrons are inelastically scattered. The dramatic decrease in image intensity in Fig. 4d compared with the (b) is mainly due to the factor $\mu_2$, indicating the strong interference of the 0.65% inelastically scattered (absorbed) electrons! This result astonishingly shows that at 20 kV, the elastic image intensity is not dominated by the number of elastically electrons contributing to the imaging, but depends on the interference between the inelastically scattered waves. The $C_c$-corrected plasmon-filtered image (Fig. 4f) conserves the elastic image contrast (Table 3), however the intensity is 39% (Table 3) of the zero-loss filtered image (Fig. 4d). The line profiles (Fig. 4g) show that intensity conservation is satisfied — the red line is the sum of the green and blue lines. In a previous paper [4], we have used the modified Weber formula to evaluate the atom contrast:

$$C_W = \frac{I_a - I_b}{I_a + I_b}.$$  

(32)

Here $I_a$ is the intensity of the atom column, and $I_b$ is the intensity of the background. This definition of the contrast is not unique. In many cases, one uses the Michelson contrast [25]

$$C_M = \frac{I_a - I_b}{I_a + I_b}.$$  

(33)

Table 3 lists the calculated atom contrast $C_W$ and $C_M$ as well as the atom intensity for the calculated images in Fig. 4. The minus sign

<table>
<thead>
<tr>
<th>UA: 50 mrad</th>
<th>Zero-loss</th>
<th>Plasmon-loss</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Without absorption</td>
<td>With absorption</td>
</tr>
<tr>
<td></td>
<td>$I_a$</td>
<td>$I_b$</td>
</tr>
<tr>
<td>47</td>
<td>1.1074</td>
<td>0.7835</td>
</tr>
<tr>
<td>48</td>
<td>1.1074</td>
<td>0.7835</td>
</tr>
<tr>
<td>49</td>
<td>1.1074</td>
<td>0.7835</td>
</tr>
<tr>
<td>50</td>
<td>1.1074</td>
<td>0.7835</td>
</tr>
</tbody>
</table>

Fig. 5. The absorption factor $\mu_2$ calculated for the two repeating patterns in (110) projection for silicon at 20 kV. As can be seen, the factor $\mu_2$ for silicon at 20 kV is close to a constant.
Table 4
Calculated image contrast and intensity for Si<110> at 20 kV. UA-usable aperture, \( I_a \)-atom column intensity, \( I_b \)-background intensity, \( C_M \)-Michelson contrast, \( C_W \)-modified Weber contrast.

<table>
<thead>
<tr>
<th>UA: 50 mrad</th>
<th>Zero-loss</th>
<th>Plasmon-loss</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Without absorption</td>
<td>With absorption</td>
</tr>
<tr>
<td>( I_a )</td>
<td>1.9525</td>
<td>1.3728</td>
</tr>
<tr>
<td>( I_b )</td>
<td>0.0562</td>
<td>0.0338</td>
</tr>
<tr>
<td>( C_M )</td>
<td>94.4</td>
<td>94.4</td>
</tr>
<tr>
<td>( C_W )</td>
<td>-3376.2</td>
<td>-3369.3</td>
</tr>
<tr>
<td>UA: 86 mrad</td>
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<td>Plasmon-loss</td>
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<tr>
<td></td>
<td>Without absorption</td>
<td>With absorption</td>
</tr>
<tr>
<td>( I_a )</td>
<td>3.0451</td>
<td>1.8275</td>
</tr>
<tr>
<td>( I_b )</td>
<td>0.0676</td>
<td>0.0407</td>
</tr>
<tr>
<td>( C_M )</td>
<td>95.7</td>
<td>95.6</td>
</tr>
<tr>
<td>( C_W )</td>
<td>-4467.4</td>
<td>-4391.5</td>
</tr>
</tbody>
</table>

Fig. 6. Simulated EFTEM images for silicon <110> at 20 kV for the SALVE II microscope (2nd column) and a microscope with an aberration corrector capable of providing an usable aperture of 86 mrad (3rd column), based on the experimental EELS spectra (1st column). Atom columns in the images are white. The 1st column: (a) The normalized zero-loss peak; (d) the zero-loss peak extracted from the normalized EELS spectra; (g) the plasmon peak. The 2nd column: the simulated EFTEM images with usable aperture 50 mrad, including the (b) zero-loss filtered image without absorption, (e) zero-loss filtered image with absorption, (h) plasmon-filtered image and (j) the line profiles marked in the image (b), (e) and (h). The 3rd column: the simulated EFTEM images with usable aperture 86 mrad, including (c) zero-loss filtered image without absorption, (f) zero-loss filtered image with absorption, (i) plasmon-filtered image and (k) the line profiles marked in the image (c), (f) and (i). Sample thickness: 16.7 ± 1 nm. Aberration parameters: \( C_C=0 \), \( C_S=0 \) and \( \Delta f=178 \text{ Å} \). Scale bar: 2 Å.

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indicates that the image of the atom is bright according to the definition of the modified Weber formula [4].

4.2. Si(110)

Si(110) has two different patterns repeating in turn along the (110) direction. Fig. 5 shows the absorption factor $\mu_2$ calculated for the two patterns. One can estimate that the factor $\mu_2$ decreases the zero-loss filtered image intensity to about $e^{-\mu_2} = e^{-0.468} \approx 0.62$, compared with the pure elastic case. The coefficients $b_0$ and $b_1$ in Eq. (25) are fitted based on the value of $\langle \mu_2 \rangle$ which lies within the range $[0, 0.5]$, in which case the linear fitting of $\exp(x) = b_0 + b_1 \cdot x$ results in $b_0 = 0.97$ and $b_1 = 1.29$, respectively. Based on the numbers and Eq. (25), one can predict that the intensity of the zero-loss filtered image calculated with mutual coherence method is $e^{-\mu_2} \cdot b_0 = 62\% \times 0.97 = 60\%$ of the intensity calculated based on pure elastic scattering, also shown by Table 4. The advantage of a large usable aperture determined by the aberration corrector is shown by comparing the second and the third column of Fig. 6, as well as the line profiles (j) and (k). As can be seen from these values, not only does the resolution increase, but also the atom column intensity increases by $(1.8275-1.1728)/1.1728 \times 100\% = 56\%$ for the usable aperture of 86 mrad compared with 50 mrad (Table 4).

With different contrast definitions, the contrast values are also different. For the usable aperture of 50 mrad and 86 mrad, the background intensities are close to 0. Therefore, the contrast defined by Michelson formula (Eq. (33)) is close to 1 in both cases. However, the contrast difference obtained by using different usable apertures is shown very pronounced by employing the modified Weber formula (Eq. (32)). The minus sign indicates that the image of the atom is bright according to the definition of the modified Weber formula [4]. The modified Weber contrast calculated for the usable aperture of 86 mrad increases by a factor of 1.3, compared with the value for the usable aperture of 50 mrad.

Intensity conservation can be verified in both cases, shown by the line profiles in Fig. 6(j) and (k) and Table 4.

5. Summary and outlook

The image simulation involving both elastic and inelastic scattering is mainly associated with two factors—$\mu_2$ and $\mu_1$. The factor $\mu_2$ accounts for the decrease of the intensity of the incident beam caused by the reduction of purely elastically scattered electrons; and the factor $\mu_1$ accounts for the increase of the intensity of the incoming beam caused by the contribution of the inelastically scattered electrons. The total intensity of the incident beam is conserved, therefore $\mu_2$ and $\mu_1$ are associated with each other, and the magnitude of both indicates how strong the interference between the inelastically scattered waves is. The calculation of $\mu_2$ and $\mu_1$ involves 4D Fourier transform of the MDFF. In order to simplify the 4D FT as combinations of 2D Fourier transforms, we have derived an approximation based on independent atom model up to an information limit of 1 Å−1 (Appendix A). Our approximation is easy to calculate and can be optimized for different imaging conditions.

We simulated EFTEM images for the SALVE II machine, based on the experimental EELS spectra obtained from the SALVE I machine. According to our simulations at 20 kV, even for an one-atom thin structure like graphene, although only less than 1% of the electrons are inelastically scattered, the strong interference of these inelastically scattered waves results in dramatic decrease of the zero-loss filtered image intensity to 72%, compared with pure elastic case. For the thicker structure Silicon (110) (17 nm thick), as

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much as 57% of the electrons are inelastically scattered, however
the zero-loss filtered image intensity drops only to 60% compared
with the pure elastic case. The examples show that especially for
low-Z materials such as carbon-based structures, the interference
between the inelastically scattered waves dominates the image
intensity. Our two examples show that in general the influence of
inelastic scattering cannot be neglected at 20 kV. The analysis on
the zero-loss and plasmon-loss filtered images shows that the
total intensity is conserved.

We may speculate that absorption caused by the interference
between inelastically scattered waves is another factor contribut-
ing to the Stobbs factor \[26\] also at higher voltages. We will report
further EFTEM experiments for graphene and silicon as soon as the
SALVE II microscope is fully operable and functional.

Moreover, we demonstrated the strong advantage of the large
usable aperture (Current Cs - CE corrector technology allows
50 mrad for our SALVE II microscope.) not only for resolution
increase, but also for contrast increase.

**Appendix A. The factorization of MDF**

We have calculated \( S(\theta, \theta', \Delta\phi) \) based on the Wentzel model
(Eq. (21)), and the factorization of \( S(\theta, \theta', \Delta\phi) \) reduces to the factorization of
\[
F(\theta - \theta') = \frac{Z \theta^2}{\theta^2 + (\theta - \theta')^2}.
\]
We introduce \( \tilde{\theta} = \theta / \theta_0 \) and temporarily leave \( Z \) out. Eq. (A.1)
becomes
\[
F(\tilde{\theta} - \tilde{\theta}') = \frac{1}{1 + (\tilde{\theta} - \tilde{\theta}')^2}.
\]
In order to approximate \( F(\tilde{\theta} - \tilde{\theta}') \) in the form of
\[
F = \sum f_j(\tilde{\theta}) g_j(\tilde{\theta}'),
\]
where \( f_j(\tilde{\theta}) \) and \( g_j(\tilde{\theta}') \) are functions depending only on \( \tilde{\theta} \) and \( \tilde{\theta}' \), respectively, we make the Ansatz
\[
F = c_1 \tilde{\omega} + c_2 \tilde{\theta} - c_3 (1 + d \tilde{\theta}^2)(1 + d \tilde{\theta}^2)
\]
Here \( c_1, c_2, c_3 \) and \( d \) are constant coefficients. In order to compare
Eqs. (A.2) and (A.3), we can discuss two cases - 1. If \( \tilde{\theta} \neq \tilde{\theta}' \), the
order of \( F \) is \(-2\), and so is the order of \( F' \). We can choose suitable
parameters of \( c_1, c_2, c_3 \) and \( d \) to minimize the difference of the two
functions. 2. If \( \tilde{\theta} = \tilde{\theta}' \), \( F \) is a constant, but the order of \( F' \) is still \(-2\).
Both \( \tilde{\theta} \) and \( \tilde{\theta}' \) plane vectors, and there are infinite positional
relations between the two vectors. The relation \( \tilde{\theta} = \tilde{\theta}' \) represents
only a special case. By choosing the proper parameters of \( c_1, c_2, c_3 \)
and \( d \), it should be possible to minimize the influence of the
unbalanced order caused by this special case.

Assuming that the angle between the two vectors \( \tilde{\theta} \) and \( \tilde{\theta}' \) is
\( \alpha \) \((0 \leq \alpha < 2\pi)\), we can write \( F \) and \( F' \) separately as
\[
F = \frac{1}{1 + \tilde{\omega}^2 - 2 \tilde{\omega} \cos \alpha}
\]
and
\[
F' = A X = \left( \frac{\tilde{\omega} \tilde{\omega} \cos \alpha}{1 + \tilde{\omega}^2} \left( \frac{1}{1 + \tilde{\omega}^2} \right) \right) c_1 c_2 c_3.
\]
Our problem becomes the minimization of the absolute difference
\( |AX - F| \). We apply the least-square method in terms of matrix. If there are \( l, j \) and \( k \) possible values for \( \tilde{\omega}, \tilde{\omega}' \) and \( \alpha \), respectively, then
\( A \) is a matrix with \( l \times j \times k \) - dimensional column space. Similarly,
\( F \) is a column vector with \( l \times j \times k \) dimensions. In order to minimize the distance \( |AX - F| \), \( AX \) needs to be the projection of
\( F \) onto the column space of \( A \). Under this condition, \( A \) should be
perpendicular to \( F - AX \), which leads to
\[
A' (F - AX) = 0
\]
\[\text{Fig. A1. Function } F \text{ and its approximation } F' \text{ for } \alpha = 0. \text{ The correlation coefficient between the approximation and the original function is } 0.9823 \text{ in this case, and the variance } \sigma^2(F - F') \text{ equals } 7.365 \times 10^{-4}. \]
the correlation between the approximated function (Eq.(A.3)) and
of the unbalanced order can be minimized by properly choosing
original function
integration of \(\cos\) is 0.9823; the mean value and the variance of \(F\)
is smaller than 0.22. One can safely assume \(\theta \approx 0\)
showing that
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